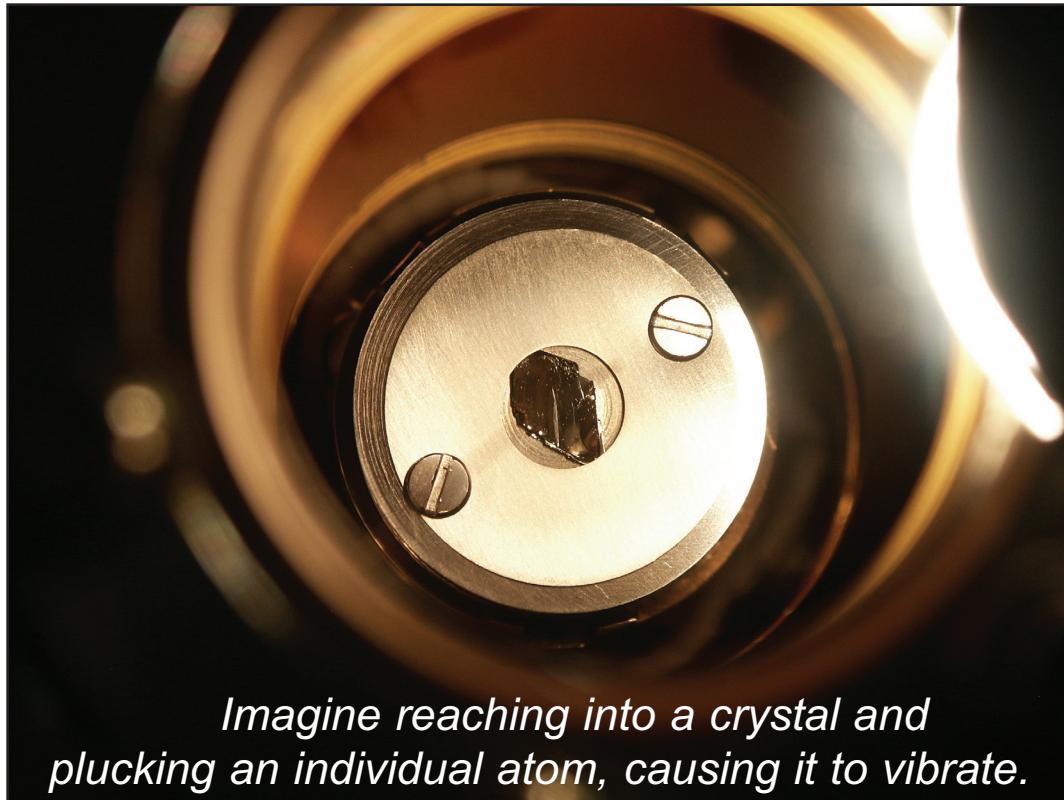


Strange atomic vibes observed in uranium at high temperatures

M.E. Manley¹



Imagine reaching into a crystal and plucking an individual atom, causing it to vibrate.

Conventional wisdom would say this vibration should spread out into the crystal, much the way water waves spread across the surface of a pond when it is disturbed. This familiar image is a natural consequence of a linear force-displacement relationship between atoms. The natural frequency of the plucked atom matches that of surrounding atoms, so it resonates with them, quickly transferring its energy in the form of waves.

About two decades ago, however, it was theorized that in the presence of non-linear forces a very different solution is possible, one in which the energy remains localized. In the presence of non-linearity the frequency of the vibrating atom depends on its amplitude, allowing the plucked atom to have a frequency distinct from all the other atoms. In this situation the vibration does not couple well to its surroundings and it becomes possible for energy to remain trapped locally for an extended period of time.

In a real crystal there is no need to pluck the atom because random thermal fluctuations will do the job. As temperature is increased configurational entropy sta-

bilizes increased concentrations of these random localized vibrations, called intrinsically localized modes (ILM), discrete breathers, or lattice solitons. This mechanism, which spontaneously concentrates energy, promises to fundamentally change our understanding of many problems in solid state physics.

Unfortunately, despite two decades of work, direct observation in a three-dimensional solid has remained elusive. Recent experiments, however, suggest that ILMs may in fact be hiding out in uranium at high temperatures.

Many of the light actinides, including uranium and plutonium, are known to be unusually non-linear at high temperatures. Raising the temperature by modest amounts leads to large changes in the frequencies of the atomic vibrations (phonons). While the underlying cause(s) for this non-linear behavior is not fully understood, it does provide the environment where ILMs might be possible. If the average frequencies change with a small temperature change it stands to reason that a thermal fluctuation can also shift the frequency locally, making possible an ILM.

¹Materials Technology: Metallurgy, Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

On reverse, the uranium crystal used in the inelastic x-ray scattering experiment.

*Photo by C.P. Opeil, S.J./
Los Alamos National Laboratory*

Michael Manley received his PhD in materials science from the California Institute of Technology in the spring of 2001 and spent a little more than a year as a Director's Postdoctoral Fellow before becoming a technical staff member in the Materials Technology and Metallurgy Group (MST-6). His current research interests are primarily experimental and extend from basic problems in metals physics to the mechanical behavior of materials.

Recent experiments suggest that intrinsically localized modes may in fact be hiding out in uranium at high temperatures.

New vibrational mode discovered

In recent measurements using both inelastic x-ray and neutron scattering, Michael Manley, MST-6, and coworkers discovered a new vibrational mode that forms in uranium at temperatures above 450 K.

The mode appears at a zone boundary, meaning that its physical size is similar to that of the crystal unit cell size, as is expected for a mode that is either intrinsically or defect localized. The appearance of the mode coincides with an anomaly in the mechanical deformation behavior but has no effect on the long range elastic properties, expected for both structural defects and ILMs. The mode appears without a change in the long range crystal structure, indicating that the required symmetry breaking must be local. The emergence of the mode is accompanied by an excess in the specific heat, implying additional entropy. This additional entropy could be accounted for by the configurational entropy of randomly distributed structural point

defects or ILMs, but not higher-dimensional defects. Dislocations, stacking faults, and other discommensurations do not contribute enough configurational entropy to stabilize themselves. Vacancies (missing atoms) are stabilized by their own entropy, but the number of vacancies needed would produce volume changes that are not seen. Impurities are also unlikely because intensity is lost in the other phonons when the new mode forms, showing that the new mode is recruiting uranium atoms that were previously involved in the normal phonons. The impurity concentrations would also have to be much higher than can be reasonably expected for either of the crystals used.

Finally, all of these structural defects would produce diffuse scattering patterns in the x-ray Laue diffraction that are not observed. These arguments, combined with the evidence of non-linear behavior, have led the researchers to conclude that the new mode is the elusive three-dimensional ILM.